

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
2 August 2001 (02.08.2001)

PCT

(10) International Publication Number
WO 01/54819 A1

(51) International Patent Classification⁷: **B02C 4/14**, 4/28,
B01J 2/14, B29B 9/08, C10L 5/02, C22B 1/245

(21) International Application Number: PCT/AU01/00062

(22) International Filing Date: 23 January 2001 (23.01.2001)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
PQ 5324 28 January 2000 (28.01.2000) AU
PQ 7872 31 May 2000 (31.05.2000) AU
PQ 9679 25 August 2000 (25.08.2000) AU

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(81) Designated States (*national*): AE, AG, AL, AM, AT, AU,
AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ,
DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR,
HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR,
LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ,
NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM,
TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

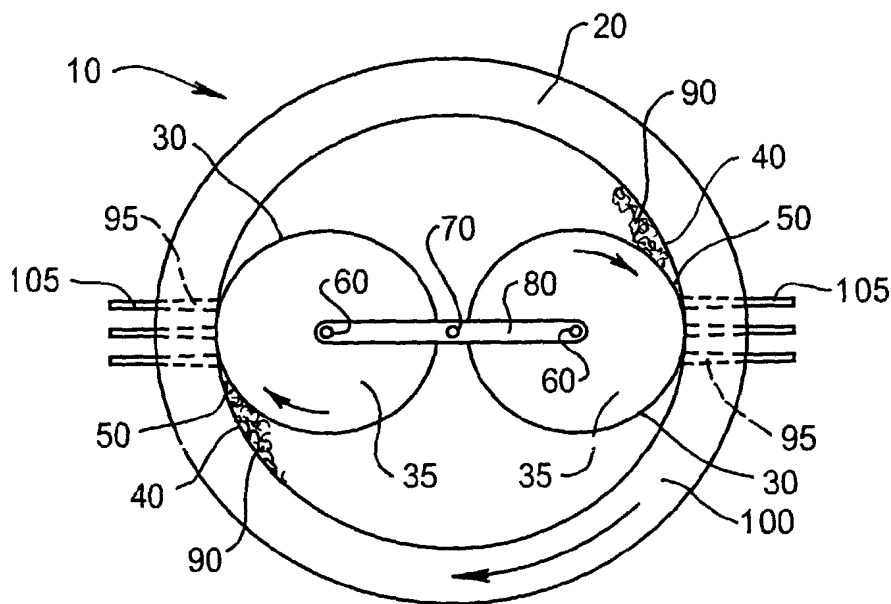
(84) Designated States (*regional*): ARIPO patent (GH, GM,
KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian
patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European
patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE,
IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF,
CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Published:

— with international search report

[Continued on next page]

(54) Title: PROCESS FOR UPGRADING LOW RANK CARBONACEOUS MATERIAL



(57) Abstract: A process for upgrading brown coal and/or the recovery of metal values. The process includes providing two or more converging surfaces defining a nip therebetween wherein, at least one of said surfaces is rollable in a direction toward said nip; feeding material to said nip where, by the rolling action of the rollable surface, under shear stresses the material forms a plastic mass. The invention also extends to a process for the production of char and/or metal char composites, with the optional recovery of metal values, by heating pellets formed from the upgraded brown coal process.

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For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

PROCESS FOR UPGRADING LOW RANK CARBONACEOUS MATERIALFIELD OF THE INVENTION

This invention relates generally to a process for upgrading low rank
5 carbonaceous material, as well as a process for recovery of metal values from
waste metal oxide particles. The invention particularly relates to an improved
process of forming pellets from low rank carbonaceous material, which pellets
may also contain waste metal oxide particles. The invention also extends to
10 processes of forming char and/or metal char composites by heat treating the
pellets, with optional recovery of metal values from the metal char composites.

BACKGROUND OF THE INVENTION

Low rank carbonaceous materials, such as brown coal, peat and lignite,
are materials having water locked into a microporous carbonaceous structure.
The water content is typically high - for example 60% or higher. This means
15 that such materials have a low calorific value. Moreover, these materials have
the undesirable mechanical properties of being soft, friable and of low density,
meaning that they are difficult, messy and inconvenient to handle.

Prior processes for upgrading low rank carbonaceous materials (which
for ease of discussion will be hereinafter collectively referred to as "brown coal")
20 have included "briquetting" and solar drying.

Briquetting typically involves heating the raw brown coal to remove
excess water, then pressing the cooled brown coal into briquettes using an
extrusion press or roll briquetting machine. However, briquetting is an
expensive process due to the requirement for thermal energy and the
25 mechanical wear on the extrusion press or roll briquetting machine.

The solar drying process involves milling of the brown coal with addition
of water for long periods (e.g. up to 16 hours), then solar drying of the milled
slurry in shallow ponds. This process is lengthy - particularly the solar drying
step which may take up to several months - and energy intensive.

30 Another proposal mechanically releases water from brown coal by
physically breaking up the coal. However, this process is inconvenient and time
consuming and still requires lengthy air drying of the final product.

It is accordingly an object of the present invention to provide a process for upgrading brown coal which overcomes, or at least alleviates, one or more disadvantages of the prior art.

SUMMARY OF THE INVENTION

5 According to the present invention, there is provided a process for upgrading brown coal as hereinbefore defined, said process including

providing two or more converging surfaces defining a nip therebetween, wherein at least one of said surfaces is rollable in a direction toward said nip;

10 feeding said brown coal to said nip where, by the rolling action of the at least one rollable surface, said brown coal is subjected to shearing stresses, causing attritioning of the microporous structure of said brown coal and the release of water contained in the micropores;

continuing the shearing attritioning until the brown coal forms into a plastic mass.

15 The present invention also provides upgraded brown coal formed from the process defined in the preceding paragraph.

The present invention further provides a process for the production of char, utilising as feed material pellets formed from the upgraded brown coal produced by the above process.

20 The present invention still further provides char produced from the process of the preceding paragraph.

Moreover, the present invention still further provides a process for recovering metal from a metal containing material, said process including:

25 providing two or more converging surfaces defining a nip therebetween, wherein at least one of said surfaces is rollable in a direction toward said nip;

30 feeding brown coal, as herein defined, and said metal containing material to said nip where, by the rolling action of the at least one rollable surface, said brown coal is subjected to shearing stresses, causing attritioning of the microporous structure of said brown coal and the release of water contained in the micropores;

continuing the shearing attritioning until the brown coal and metal containing material form into a composite plastic mass;

heating the composite mass in order to pyrolyse the brown coal and form sufficient reductant to reduce said metal containing material to said metal,

thereby producing a reduced composite including said reduced metal and a carbonaceous phase.

The present invention also provides a composite of metal containing material and upgraded brown coal formed according to the above process.

- 5 Further, the present invention provides a reduced composite formed according to the above process.

DETAILED DESCRIPTION OF THE INVENTION

- Accordingly, the brown coal is comminuted by a process involving shearing attritioning as distinct from simple grinding. Typically this process
10 takes place in a mill. Preferably the mill includes at least one roller. Preferably, the mill does not include an air separator as such a device is not compatible with the process of the invention and can interfere with the shearing/attritioning and/or extrusion of the plastic mass.

- It is believed that the rolling action of the at least one converging surface
15 towards said nip is advantageous because the brown coal is actively directed into the nip and subjected to more efficient shearing forces there than would be the case using, for example, a blending or kneading apparatus simply having rotating paddles. In such an apparatus, shearing stresses are produced in a narrow gap between the walls of the blender and the rotating paddles and are
20 generally not as efficient as the shearing stresses generated during the process of the present invention.

The shearing attritioning may be preceded, if necessary, by a size reduction step in which the brown coal is subjected to grinding, such as in a hammer mill.

- 25 The shearing attritioning is preferably effected in a rotating roll type pelletising mill. Such a mill typically comprises a housing, preferably drum or cylindrical in shape, within which is provided at least one rollable curved surface, typically the surface of a roll, preferably a cylindrical shaped roll. The curved inner surface of the housing and the curved surface of, for example, the
30 roll are positioned relative to each other so as to provide two converging surfaces between which is defined a nip. This typically will require that the axis of rotation of the roll will be eccentric relative to the axis of rotation of the housing. In use, there is relative rotational movement between the two surfaces. This may be effected by rotation of the housing about its axis and/or

rotation of the roll about its axis. Alternatively, the housing may be stationary and the roll rotates about the axis of the housing as well as rotating about its own axis. The rotation of the roll about the axis of the housing may be effected by means of an arm mounted for rotatable movement at the axis of the housing, and having at one end thereof the roll also mounted for rotatable movement. The relative rotational movement of the two surfaces is such as to cause the brown coal to be forced into the nip, where it is subjected to shearing attritioning.

In one preferred embodiment of the invention, an arm is mounted for rotatable movement at the axis of the housing and a roll is mounted at either end of the arm. In such an arrangement the mill has in effect two pairs of converging surfaces, with each roll providing a rollable surface defining a nip where the rollable surface is closest to the inner surface of the housing.

It is believed that the shearing attritioning of the coal particles causes breakage of bonds between coal particles with consequential release of water trapped in micropores of the coal structure.

Preferably, the attritioned carbonaceous material is then subjected to extrusion which further shears the material. Preferably the extrusion process occurs substantially immediately after or concurrently with the shearing attritioning. Most preferably the shearing attritioning and extrusion occur in a single operation, typically in a single apparatus, which avoids the need to transfer the material from an attritioning apparatus to an extruder. The extrusion is advantageously effected by forcing the attritioned material through tapered apertures, having decreasing diameter as the material is pushed through. The tapered apertures effect the application of very high pressures to the material during extrusion, causing further mechanical release of water from the micropores of the brown coal and forcing the coal particles into close proximity thereby promoting renewed bonding between the particles. The apertures typically have a diameter ranging from about 8 to about 20 mm, preferably about 8 to about 15 mm, more preferably from about 10 to about 12 mm. The length of the apertures typically range from about 15 to about 100 mm, preferably from about 30 to about 90 mm, more preferably from about 30 to about 60 mm.

In another type of rotating roll type pelletising mill, one or more rollable curved surfaces are positioned closely adjacent a substantially planar surface, with each rollable curved surface together with the planar surface defining a nip therebetween. The rollable curved surface is typically the surface of a cylindrical shaped roll. Preferably, the axis of rotation of each roll is essentially parallel with the planar surface. Typically the or each cylindrical roll is positioned for rotatable movement closely above the planar surface. The or each rotatable roll may additionally be mounted for rotatable movement about an axis perpendicular to the planar surface. The attritioned brown coal is then subjected to extrusion by being forced through apertures in a die, the upper surface of which forms the planar surface. Cylinders of extruded brown coal exit the other side of the die where they are cut into pellets.

Surprisingly the inventors have discovered that rotating roll type pelletising mills, which are commonly used in the production of pelleted animal feed, fertilisers and pharmaceuticals, or in the densification of dusty materials, are unexpectedly suitable for use in the process of the invention. For example, suitable machines are those sold by the company Sprout Waldron & Company, Inc. in Muncy, Pennsylvania 17745, United States of America, such as the pellet mill having the trade name Sprout Waldron Junior Ace, and those sold by California Pellet Mill. Further, suitable machines also include flat die pelleting presses, such as those sold by Amandus Kahl GmbH & Co.

In using a rotating roll type pelletising mill, such as the Sprout Waldron pelleting mills, the brown coal is subjected to shearing attritioning at the nip between the surface of each roll and another surface in the mill. That other surface also forms part of an extruder die, having holes through which the attritioned brown coal is extruded. Accordingly, simultaneously with the shearing attritioning at each nip, the attritioned brown coal is forced through the holes of the die by the action of the roll. The attritioned brown coal is thereby compressed into solid cylinders which are cut into pellets as they emerge by cut-off knives. The combined operations of shearing, attritioning and extrusion occur in a very short time period (e.g. fractions of a second), thus avoiding lengthy time periods which would otherwise be required to first form an extrudable paste and then transfer the paste to an extruder from which pellets are produced.

In the Sprout Waldron pelleting mill, shearing attritioning occurs at the nip between the surface of each roll and the inner wall of the rotating housing. The housing is also an extruder die having tapered holes therethrough with decreasing diameter from the inner wall to the outer wall. Tapered holes ensure the application of very high pressures to the material during extrusion. A rotatable arm is mounted for rotatable movement about the axis of the housing and a roll is mounted for rotatable movement at each end of the arm. The rotatable arm is typically powered by a 50 hp motor. The diameter of the tapered holes in the die can be as small as 10 to 12 mm. It will therefore be appreciated that an extremely efficient shearing force is exerted on the brown coal at the nip and that it is subjected to very high pressure as it is forced through the tapered holes, thereby forcing the coal particles into very close proximity with each other. Accordingly, water loss from the microstructure and renewed bonding between coal particles is maximised. The act of extrusion also substantially increases the temperature of the emerging pellets, which may be as high as 50°C. Such a high temperature enhances evaporation of surficial water from the pellets released from the micropores. This feature of the process of the invention is extremely advantageous since it ensures substantial water loss from the pellets in the very early stage of the drying process, thereby considerably minimising the overall drying time.

In some circumstances, pelletising machines sold by Warren & Baerg Manufacturing Inc., such as the Model 250 Cuber, may be used. However due to the less effective shearing action of this machine, it may be necessary to additionally use a further attritioner and/or extruder in combination with it.

The process of the invention may advantageously be used to produce feed material for use in a subsequent process for the production of char.

Alternatively, the process is particularly advantageous in the production of composites of attritioned brown coal with a metal containing material. These composites can be subsequently used as feed material in a process for recovery of the metal, in which the composite pellets are heated to a sufficiently high temperature to effect pyrolysis of the brown coal and consequent reduction of the metal containing material (typically metal oxide or sulphide) to its metal. The sheared and attritioned brown coal used in such composites exhibits an enhanced reducing potential compared with a brown coal which has been

comminuted by simple grinding. As a result, reduction of the metal containing material can advantageously take place at temperatures significantly lower than conventional reduction temperatures.

Accordingly, significant advantages can be achieved by shearing
5 attritioning of the brown coal, which are not widely recognised. Where shearing attritioning is conducted to upgrade brown coal per se, or to produce feed for char production, more highly efficient water removal is achieved and the subsequent pyrolysis of brown coal may occur at a lower than conventional pyrolysis temperature. Furthermore, shear attritioned brown coal in composites
10 of brown coal and metal containing material has a substantially increased reduction potential, as compared with brown coal which has been comminuted by simple grinding.

DESCRIPTION OF DRAWINGS

The invention will become more readily apparent from the following
15 exemplary description in connection with the accompanying drawings and Examples.

Figure 1 is a side-on view of a first pelletising mill suitable for use in the method of the invention.

Figure 2 is a perspective view of a second pelletising mill suitable for use
20 in the method of the invention.

Two different embodiments of pelletising mills suitable for use in the method of the invention are illustrated schematically in Figures 1 and 2.

One type of rotating roll type pelletising mill 10, illustrated schematically in Figure 1, comprises a housing 20, having a cylindrical shape, within which is
25 provided two rollable curved surfaces 30, each comprising the surface of a cylindrical shaped roll 35. The curved inner surface 40 of the housing 20 and the curved surface 30 of each roll are positioned relative to each other so as to provide two converging surfaces between which is defined a nip 50. This requires that the axis of rotation 60 of the roll 35 be eccentric relative to the axis
30 of rotation 70 of the housing 20. In use, there is relative rotational movement between the two surfaces 30,40 in order to effect the rolling action. This may be effected by rotation of the housing 20 in the direction of the arrow about its axis and/or rotation of each roll 35 about its axis 60, also as indicated by the direction of the arrow. Alternatively, the housing 20 may be stationary and each

roll 35 rotates about the axis 70 of the housing 20 as well as rotating about its own axis 60. The rotation of each roll 35 about the axis 70 of the housing 20 is effected by means of an arm 80 mounted for rotatable movement at the axis 70 of the housing 20. A roll 35 is mounted at either end of the arm 80. In such an arrangement the mill has in effect two pairs of converging surfaces, with each roll 35 providing a rollable surface 30 defining a nip 50 where the rollable surface 30 is closest to the inner surface 40 of the housing 20.

The attritioned carbonaceous material 90 is then subjected to extrusion which further shears the material. The shearing attritioning and extrusion occur in a single operation which avoids the need to transfer the material from an attritioning apparatus to an extruder. The extrusion is effected by forcing the attritioned material 90 through tapered apertures 95, having decreasing diameter as the material is pushed through. For simplicity, only a few of the apertures are depicted in Figure 1 whereas in fact the apertures 95 extend substantially completely around the entire housing 20, such that the housing 20 functions as a die 100. The tapered apertures 95 effect the application of very high pressures to the material during extrusion, causing further mechanical release of water from the micropores of the brown coal and forcing the coal particles into close proximity thereby promoting renewed bonding between the particles. The extruded material forms into cylinders 105 which may be cut into pellets.

In another type of rotating roll type pelletising mill 110, illustrated schematically in Figure 2, one or more rollable curved surfaces 130 are positioned closely adjacent a substantially planar surface 140, with each rollable curved surface 130 together with the planar surface 140 defining a nip 150 therebetween. The rollable curved surface 130 is the surface of a cylindrical shaped roll 135. The axis of rotation X,Y of each roll 135 is essentially parallel with the planar surface 140. Each cylindrical roll 135 is positioned for rotatable movement closely above the planar surface 140. Each rotatable roll 135 is additionally mounted for rotatable movement about an axis A perpendicular to the planar surface 140. The attritioned brown coal 190 is subjected to extrusion by being forced through apertures 195 in a die 200, the upper surface of which forms the planar surface 140. Cylinders of extruded brown coal 205 exit the other side of the die 200 where they can be cut into pellets.

The pellets produced by the process of the invention can advantageously be used as feed material for subsequent production of carbon char. The pellets are typically fed to a kiln or a retort, preferably a rotary kiln. Typically, the rotary kiln comprises an inclined, cylindrical furnace which rotates slowly about its axis. The pellets are fed into the upper end and they move slowly through to the lower end under the influence of gravity and with the assistance of spiral baffles inside the furnace. While the initial start up of the kiln is effected using an external source of fuel (such as natural gas, oil or pulverised coal) combustible gases given off the pellets during the process enable the process to be subsequently self fuelling.

The brown coal containing pellets typically contain around 12% total water, plus carbon, volatiles and minerals derived from the original brown coal. During heating of the pellets, free water is first evolved, followed by combined water once the temperature reaches about 250°C. With increasing temperature, volatiles, mainly hydrocarbons such as methane, are released between 400 and 700°C. These volatiles largely decompose to carbon monoxide, hydrogen and some carbon dioxide. The product of the charring process is the original carbon plus ash derived from the minerals.

It is an advantage of the present invention that the pellets produced from the inventive shearing attrition and extrusion process are sufficiently dense and strong that they can be successfully used as feed material in a rotary kiln without significant break up. This is a considerable advance over prior art briquettes which tend to break when processed in a rotary kiln.

In an alternative embodiment of the process of the invention, metal value containing material can be combined with the brown coal during shearing attritioning and incorporated in the subsequently produced pellets. The production of such composite pellets may be desirable in order to convert difficult to handle, fine metal oxide dusts, such as that produced as waste from electric arc furnaces, into a more convenient and easy to handle form. Such pellets can also be used as feed material in a subsequent process to reduce and recover the metal values.

This modification of the inventive process will now be described with particular emphasis on its application to the recovery of metal values from dusts generated from the melting of iron and steel, particularly the melting of steel

scrap in electric arc furnaces. However, it is to be understood that the process is not limited to such use, and can be generally applied to recovery of metal values from any materials, particularly particulate materials. Other metal containing material may include copper containing material, such as copper sulphides and/or copper oxides.

Dusts generated during the melting of iron and steel, more particularly the melting of steel scrap in electric arc furnaces, commonly contains a good deal of zinc (say 20 to 30%) together with an amount of iron and much smaller quantities of lead and various other minor elements. The safe disposal of such material presents substantial difficulties because, if used in land fill operations soluble elements such as zinc, cadmium and copper may readily leach into the ground water. If stockpiled such material is subject to wind dispersal and the considerable quantities generated render such an approach impractical. In some current operations the dust is transported to separate fuming plants for treatment but this is costly and results in the recovery of only a proportion of the contained zinc and none of the iron.

The process of the invention enables the recovery of very high proportions of all elements involved in economic forms without leaving any hazardous residues. Moreover, the treatment plants may be small in size, not capital intensive and may be located conveniently near to the electric arc furnace plants mentioned above.

The proposed process uses low rank carbonaceous materials, such as brown coal, peat, soft lignites of relatively high water content or plant derived materials either singly or in combination in thorough mixture with the dust in the form of composite pellets or agglomerates using the process of the invention.

As previously described, brown coal releases water contained in its microstructure when shear attritioned to form a wet, smooth plastic mass, then the coal particles rapidly rebond with substantial loss of water to form a relatively hard dense product of low water content. The electric arc furnace dust added to the coal prior to shearing attritioning becomes incorporated into the final hard product in very close association with the coal particles.

The composite pellets thus produced can be used as feed material in a subsequent hot briquetting process such as to form reduced carbon/iron

briquettes and/or in a process to recover metal values from the electric arc furnace dust.

When the hard dry pellets are heated in, for instance, a semi-closed retort or kiln, preferably a rotary kiln, to maintain the reducing atmosphere generated by the coal, the pyrolysing brown coal can exert an extremely strong reducing effect at quite low temperatures (500 - 700°C). The finely particulate metal oxides in any furnace dust contained within the composites are thus exposed to the full reducing action and can be rapidly and completely reduced to the metallic state. This applies not only to the zinc, lead, copper and cadmium but also to the iron which is a major component of the dust.

Whilst not wishing to be restricted to a particular reduction mechanism, when iron oxides are present in the composite pellet, as the kiln temperature rises above 500°C, it is believed that reduction of hematite (Fe_2O_3) to magnetite (Fe_3O_4) occurs and, in turn, reduction of the latter to wustite (FeO) then occurs. Subsequently, wustite is reduced to metallic iron from around 900°C. During pyrolysis of the brown coal, sufficient volatiles are preferably produced to satisfy the demand for chemical reductant as well as to provide at least most of the fuel for the process. The reduced pellets therefore contain Direct Reduced Iron (DRI), which is a generic term for the product of the process in which oxidic iron ores or concentrates are reduced to metallic iron in their solid state by reductants such as hydrogen, carbon monoxide and coal.

An advantage of the invention is that the temperature of reduction of the iron oxide in the composite pellets takes place around 500-700°C, which is considerably lower than the reduction temperature ordinarily required, about 1200°C.

The reduced pellets may be further processed by hot or cold briquetting possibly after crushing. Where the reduced pellets containing DRI are subjected to hot briquetting, this results in Hot Briquetted Iron (HBI). This product typically takes the form of dense, hard pellets containing intimately mixed metallic iron and carbon. The hot forming process densifies material, e.g. by reducing porosity, and excludes air and/or moisture, thereby minimising oxidation of iron and carbon in the briquettes. The HBI can be stock piled and may subsequently be used as feedstock in furnaces for making iron and/or steel.

An advantageous feature of the present invention is that the reduced pellets may alternatively be subjected to cold briquetting. In this process, the reduced pellets are allowed to cool in an inert atmosphere to about ambient temperature before being formed into briquettes. The cooling in the presence of an inert atmosphere minimises oxidation of iron and carbon in the pellets prior to their formation into briquettes. The pellets may be crushed and may be blended with a binder or other additive before briquetting. As for hot briquetting, cold briquetting densifies the material by reducing porosity and excludes air and moisture to minimise oxidation of iron and carbon.

Subsequent to, or instead of, hot or cold briquetting, the process may further include separation of the reduced metals from the residual char and gangue. Where the pellets include reduced metals derived from electric arc furnace dusts, that separation may be by one of two options.

In the first option, the metallic zinc, lead and cadmium recovered are volatilised and the vapours are conducted to a suitable condenser with the aid of an inert carrier gas, e.g. nitrogen, argon. This is achieved by raising the temperature progressively to about 1000°C until complete removal of the volatile metals has taken place. The residual composites will then contain only reduced iron, carbon and gangue and may be returned to the furnace. Care must be taken throughout the process to ensure that no oxidising gases are present during volatilisation.

The second, preferred alternative is to produce zinc oxide and melted iron directly from the hot reduced composites. For this purpose the reduced composite pellets, at about 700°C may be discharged into a suitable vessel with provision for bottom blowing and oxygen injection at an appropriate rate. Combustion of the hot char will cause a rapid rise in temperature with volatilisation of the zinc, lead and cadmium which will then oxidise in the atmosphere above the charge. The oxidised metals may readily be collected in a bag house system. The bulk of the residual iron in the burning composite will rapidly melt, thus permitting effective recovery contained in the original furnace dust.

An appreciation of the volatilities of the base metals concerned will assist an understanding of the principles underlying the process of the invention. The boiling points of zinc, cadmium and lead are set out in Table 1.

TABLE 1

| | <u>Metal</u> | <u>Boiling Point °C</u> |
|---|--------------|-------------------------|
| | Zinc | 906 |
| | Cadmium | 765 |
| 5 | Lead | 1740 |

In the first stage of pyrolysis/reduction of the composites the temperature should be limited to about 700 - 725°C so as to avoid volatilisation of the zinc and cadmium. This temperature range is sufficient to achieve full reduction of the metal oxides and removal of most of the organic volatiles (mainly phenols) from the heated coal.

It should be noted that although lead is relatively non-volatile that there is some carry-over of this metal during heating to 1000°C in a carrier gas stream - presumably in the form of micro-droplets.

EXAMPLES

The process of the invention will be better understood by reference to the following non-limiting Examples.

Example 1 : Formation of Upgraded Brown Coal Pellets

Loy Yang brown coal having 60% by weight water as mined, reducing to 55% by weight water after storage in bags, was subjected to shearing attritioning and extrusion in a Sprout-Waldron Junior Ace pelleting mill. The wet pellets as formed had a diameter of 12 mm. The pellets were allowed to dry naturally in an open shed with free air movement. The maximum day temperature during the trial was 26 to 28°C. The water content of the pellets was measured over time and is presented in Table 2.

TABLE 2

| | <u>Time</u> | <u>Water %</u> |
|---|--|----------------|
| | Before pelletising | 55 |
| | 15-20 minutes after formation | 38 |
| 5 | 1 hour after pellet formation | 36.5 |
| | 4 hours after pellet formation | 23.0 |
| | 20 hours after pellet formation (next day) | 17.5. |

These results illustrate the very rapid water loss and accompanying densification hardening attainable using the procedure of this invention. After 10 20 hours the above pellets were suitable for subsequent applications.

Examples 2 and 3 : Recovery of Metal Values from Composite Pellets of Brown Coal and Electric Arc Furnace Dust

In each case brown coal from Morwell, Victoria together with electric arc furnace dust from Smorgon Steel, Laverton, Victoria were used. The dust had 15 the following compositions as set out in Table 3.

TABLE 3

| | <u>Element</u> | <u>% by weight</u> |
|----|---|--------------------------|
| | Zinc | 27.45 |
| | Iron | 21.08 |
| 20 | Calcium | 3.49 |
| | Lead | 2.36 |
| | Manganese | 1.16 |
| | <u>Together with</u> the minor elements | |
| | | <u>Parts per million</u> |
| 25 | Copper | 1730 |
| | Cadmium | 375 |
| | Nickel | 206 |
| | Cobalt | 5. |

Example 2

30 Composite pellets made in accordance with the process of the invention and having a coal:dust ratio of 1:2 were used as feed to a retort furnace. The retort was heated to about 700°C over fifty minutes and the temperature then raised progressively first to 940°C and finally to 1050°C over a thirty minute period. During the latter stages of heating a stream of pre-heated oxygen free

nitrogen was used to carry the volatile metals out of the retort to a simple tube condenser isolated from the atmosphere by a water trap. The efficiency of the process in this example was determined by analysis of four separate samples of the residual cooled char composites in the retort. The results are presented in

5 Table 4.

TABLE 4

| | <u>Sample</u> | <u>% Zinc Volatilised</u> |
|----|---------------|---------------------------|
| | 1 | 97.52 |
| | 2 | 97.27 |
| 10 | 3 | 97.67 |
| | 4 | 97.51. |

The condensed metal had the following composition as set out in Table

5.

TABLE 5

| | <u>Element</u> | <u>% by weight</u> |
|----|----------------|--------------------|
| 15 | Zinc | 99.3 |
| | Lead | 0.50 |
| | Iron | 0.12 |
| | Magnesium | 0.0003 |
| 20 | Manganese | 0.002 |
| | Copper | 0.0001 |
| | Cadmium | 0.0025 |
| | Nickel | 0.024 |
| | Cobalt | 0.001. |

25 Example 3

Composite pellets made according to the process of the invention and having a coal:dust ratio of 1:1 were used as feed to a retort furnace. The retort was heated to about 700°C over 60 minutes and the temperature was then raised to 1170°C over a further 80 minutes, the second heating stage being
 30 accompanied by a stream of nitrogen through the retort to carry the volatilised metal to the condenser. Efficiency was determined by measuring both recovered metal (zinc) in the condenser and residual zinc in the cooled char. Four separate determinations indicated 100% volatilisation.

The composition of the condensed metal is set out in Table 6.

TABLE 6

| | <u>Element</u> | <u>% by weight</u> |
|----|----------------|--------------------|
| | Zinc | 97.3 |
| | Lead | 1.32 |
| 5 | Iron | 0.32 |
| | Manganese | 0.058 |
| | Copper | 0.006 |
| | Cadmium | 0.175 |
| | Nickel | 0.0026 |
| 10 | Cobalt | 0.001. |

Analysis of the condensed metal indicated rather more transport of lead than might be expected from the higher final temperature.

The preceding Examples demonstrate that the process of the invention enables virtually complete separation and recovery of zinc from furnace dusts.

15 Examples 4 and 5 : Preparation of Cold Briquettes

Example 4

Example 4 demonstrates the preparation of briquettes by compaction of cold sponge iron (DRI) prepared by the reduction of iron oxides in composite pellets prepared according to the invention and allowed to cool to ambient
20 temperature.

45 g of cold sponge iron was blended with 1.0 g of a binder and 1.0 ml of water and compacted using a load of 48 tonnes (264 MPa). The resultant cold briquette required only limited air drying prior to utilisation as feedstock for further processing at ambient temperature.

25 Example 5

Example 5 further demonstrates the method of preparation of cold briquetted sponge iron prepared by the reduction of iron oxides in composite pellets formed according to the invention.

45 g of cold sponge iron and impurity phases were crushed to a uniform
30 mixture and blended with 1 g of a binder, in this case stearic acid. The resultant mixture was compacted using a load of 30 tonnes (165 MPa). The resultant briquettes required no further processing prior to use as feedstock for the production of iron.

Further it is to be understood that various alterations, modifications and/or additions may be introduced into the constructions and arrangements of steps previously described without departing from the spirit or ambit of the invention.

CLAIMS:

1. A process for upgrading brown coal, as hereinbefore defined, said process including:
 - 5 providing two or more converging surfaces defining a nip therebetween, wherein at least one of said surfaces is rollable in a direction toward said nip; feeding said brown coal to said nip where, by the rolling action of the at least one rollable surface, said brown coal is subjected to shearing stresses, causing attritioning of the microporous structure of said brown coal and the
10 release of water contained in the micropores; and continuing the shearing attritioning until the brown coal forms into a plastic mass.
2. The process of claim 1, wherein said process takes place in a mill
15 including at least one roller, preferably in a rotating roll type pelletising mill.
3. The process of claim 2, wherein said mill does not include an air separator.
- 20 4. The process of any preceding claim, including the further step of extruding the attritioned brown coal either substantially immediately after or concurrently with the shearing attritioning.
5. The process of claim 4, wherein the shearing attritioning and extruding
25 steps occur in a single operation, preferably in a single apparatus.
6. The process of claim 4 or 5, wherein said extruding is effected by forcing the attritioned brown coal through apertures by the rolling action of the rollable surface.
30
7. The process of claim 6, wherein said apertures have decreasing diameter as the attritioned brown coal is pushed therethrough.

8. The process of claim 6 or 7, wherein said apertures have a diameter ranging from about 8 to about 20 mm and a length ranging from about 15 to about 100 mm.

5 9. The process of any one of claims 6 to 8, wherein said apertures are provided in at least one of said converging surfaces.

10. The process of any one of claims 4 to 9, wherein the extruded attritioned brown coal is cut into pellets.

10

11. The process of claim 10, wherein said pellets are used as feed material in a process for the production of char.

12. The process of claim 11, wherein said process for the production of char
15 is conducted in a rotary kiln.

13. The process of claim 11 or 12, wherein during said process for the production of char, pyrolysis of the brown coal in the pellets takes place between 400 and 700°C.

20

14. The process of any one of claims 1 to 10, wherein a metal containing material, such as dust generated from the melting of iron and steel, is mixed with said brown coal prior to or during the shearing attritioning to form a composite plastic mass.

25

15. A process for recovering metal from a metal containing material, said process including:

providing two or more converging surfaces defining a nip therebetween, wherein at least one of said surfaces is rollable in a direction toward said nip;

30

feeding brown coal, as herein defined, and said metal containing material to said nip where, by the rolling action of the at least one rollable surface, said brown coal is subjected to shearing stresses, causing attritioning of the microporous structure of said brown coal and the release of water contained in the micropores;

continuing the shearing attritioning until the brown coal and metal containing material form into a composite plastic mass;

heating the composite mass in order to pyrolyse the brown coal and form sufficient reductant to reduce said metal containing material to said metal,
5 thereby producing a reduced composite including said reduced metal and a carbonaceous phase.

16. The process of claim 15, wherein said metal containing material is a metal oxide or sulphide, such as an oxide or sulphide of iron, zinc, lead, copper
10 or cadmium.

17. The process of claim 15 or 16, wherein the metal containing material is an oxide or sulphide of iron or copper.

15 18. The process of any one of claims 15 to 17, wherein said metal containing material is dust generated from the melting of iron and steel, preferably dust generated from melting steel scrap in an electric arc furnace.

19. The process of any one of claims 15 to 18, including the further step prior
20 to said heating step of extruding the composite plastic mass either substantially immediately after or concurrently with the shearing attritioning.

20. The process of claim 19, wherein the shearing attritioning and extruding steps occur in a single operation, preferably in a single apparatus, more
25 preferably in a rotating roll type pelletising mill.

21. The process of claim 20, wherein said mill does not include an air separator.

30 22. The process of any one of claims 19 to 21, wherein said extruding is effected by forcing the composite mass through apertures by the rolling action of the rollable surface.

23. The process of claim 22, wherein said apertures have decreasing diameter as the composite mass is pushed therethrough.

24. The process of claim 22 or 23, wherein said apertures have a diameter
5 ranging from about 8 to about 20 mm and a length ranging from about 15 to about 100 mm.

25. The process of any one or claims 22 to 24, wherein said apertures are provided in at least one of said converging surfaces.

10

26. The process of any one of claims 19 to 25, wherein the extruded composite mass is cut into pellets prior to said heating step.

27. The process of any one of claims 15 to 26, wherein said heating step is
15 conducted in a semi-closed retort or kiln, preferably a rotary kiln.

28. The process of any one of claims 15 to 27, wherein the pyrolysis of the brown coal in said heating step takes place at a temperature between 500 and 700°C.

20

29. The process of any one of claims 18 to 28, wherein said dust includes iron oxide and the reduced metal formed by reducing the iron oxide in the heating step is Direct Reduced Iron (DRI).

25 30. The process of claim 29, wherein said DRI is subjected to hot or cold briquetting, preferably cold briquetting.

31. The process of claim 30, wherein prior to said cold briquetting, said DRI is cooled in an inert atmosphere to about ambient temperature.

30

32. The process of any one of claims 15 to 31, wherein said reduced metal is separated from the reduced carbonaceous phase in said reduced composite.

33. The process of any one of claims 29 to 31, wherein said reduced composite is heated in the presence of oxygen in order to combust the reduced carbonaceous phase and increase the temperature of the reduced composite sufficiently to melt the iron in the composite.

5

34. Upgraded brown coal formed according to the process of any one of claims 1 to 10.

35. Char produced according to the process of any one of claims 11 to 13.

10

36. A composite of metal containing material and upgraded brown coal formed according to the process of claim 14.

37. A reduced composite formed according to the process of any one of claims 15 to 31.

15

38. Metal formed according to the process of any one of claims 32 and 33.

39. A process for upgrading brown coal, substantially as herein described with reference to the accompanying drawings.

20

40. A process for upgrading brown coal, substantially as herein described with reference to Example 1.

41. A process for recovering metal from a metal value containing material, substantially as herein described with reference to the accompanying drawings.

25

42. A process for recovering metal from a metal value containing material, substantially as herein described with reference to any one of Examples 2 to 5.

30

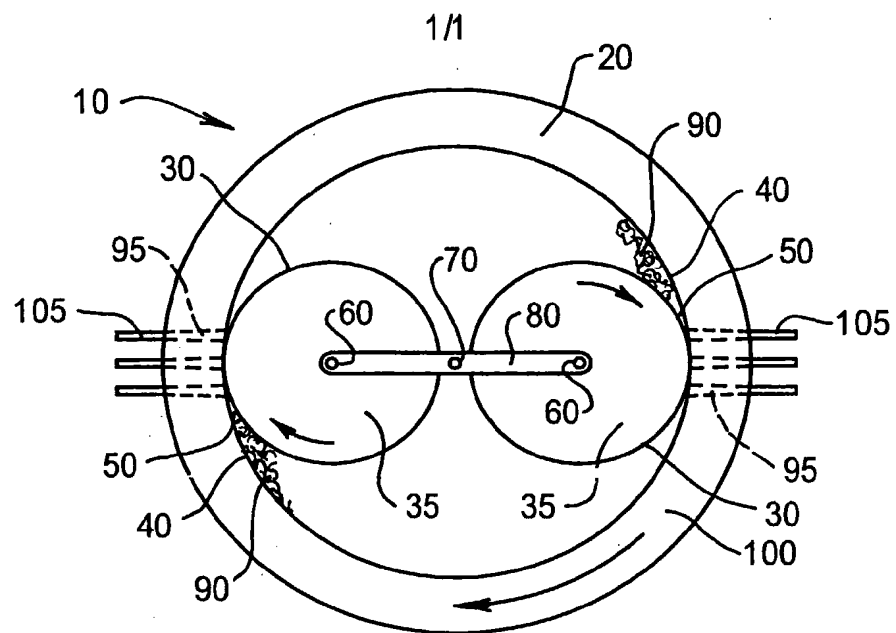


FIG 1

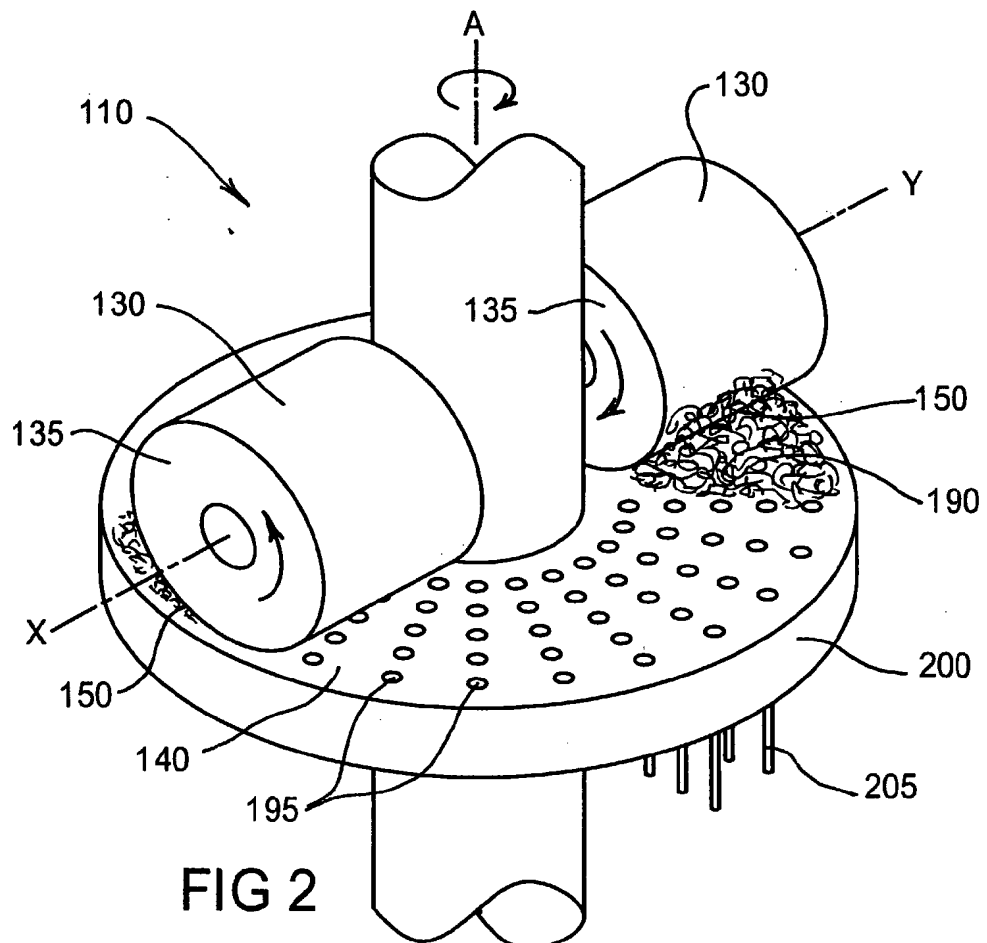


FIG 2

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU01/00062

| | | | | | | | | | | | | | | |
|---|--|---|--|---|--|--|---|--|---|---|--|--|--|--|
| A. CLASSIFICATION OF SUBJECT MATTER | | | | | | | | | | | | | | |
| Int. Cl. ⁷ : B02C 4/14, 4/28; B01J 2/14; B29B 9/08; C10L 5/02; C22B 1/245 | | | | | | | | | | | | | | |
| According to International Patent Classification (IPC) or to both national classification and IPC | | | | | | | | | | | | | | |
| B. FIELDS SEARCHED | | | | | | | | | | | | | | |
| Minimum documentation searched (classification system followed by classification symbols) | | | | | | | | | | | | | | |
| IPC B02C 4/10, 4/12, 4/14, 4/28; B01J 2/14, 2/22; B29B 9/08; C10L 5/02, 5/04; C22B 1/14, 1/245 | | | | | | | | | | | | | | |
| Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched | | | | | | | | | | | | | | |
| AU:IPC as above | | | | | | | | | | | | | | |
| Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) | | | | | | | | | | | | | | |
| WPAT: (coal+ or peat+ or lignit+) or ((coal+ or carbonac+) and pellet+) | | | | | | | | | | | | | | |
| C. DOCUMENTS CONSIDERED TO BE RELEVANT | | | | | | | | | | | | | | |
| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. | | | | | | | | | | | | |
| X | GB 2 172 586 A (CRA Services Limited) 24 September 1986 see page 1 lines 5 to 115 | 1-6, 10-22, 26-29, 32, 34-38 | | | | | | | | | | | | |
| X | US 4 758 244 A (Harvey et al.) 19 July 1988, see column 1 lines 10 to 11, column 2 lines 11 to 39, columns 5 to 6, and claims 1-5 | 1-6, 10-15, 27-28, 32, 34-38 | | | | | | | | | | | | |
| X | AU 11627/83 A (Idemitsu Kosan Company Limited) 22 September 1983 see pages 2 to 3 | 1-6, 10-13, 34-35 | | | | | | | | | | | | |
| <input checked="" type="checkbox"/> Further documents are listed in the continuation of Box C <input checked="" type="checkbox"/> See patent family annex | | | | | | | | | | | | | | |
| <table border="0"> <tr> <td>* Special categories of cited documents:</td> <td>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</td> </tr> <tr> <td>"A" document defining the general state of the art which is not considered to be of particular relevance</td> <td>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</td> </tr> <tr> <td>"E" earlier application or patent but published on or after the international filing date</td> <td>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</td> </tr> <tr> <td>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</td> <td>"&" document member of the same patent family</td> </tr> <tr> <td>"O" document referring to an oral disclosure, use, exhibition or other means</td> <td></td> </tr> <tr> <td>"P" document published prior to the international filing date but later than the priority date claimed</td> <td></td> </tr> </table> | | | * Special categories of cited documents: | "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention | "A" document defining the general state of the art which is not considered to be of particular relevance | "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone | "E" earlier application or patent but published on or after the international filing date | "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art | "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) | "&" document member of the same patent family | "O" document referring to an oral disclosure, use, exhibition or other means | | "P" document published prior to the international filing date but later than the priority date claimed | |
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| "O" document referring to an oral disclosure, use, exhibition or other means | | | | | | | | | | | | | | |
| "P" document published prior to the international filing date but later than the priority date claimed | | | | | | | | | | | | | | |
| Date of the actual completion of the international search 14 March 2001 | | Date of mailing of the international search report 21 March 2001 | | | | | | | | | | | | |
| Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA E-mail address: pct@ipaustalia.gov.au Facsimile No. (02) 6285 3929 | | Authorized officer JOHN DEUIS Telephone No : (02) 6283 2146 | | | | | | | | | | | | |

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU01/00062

| C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT | | |
|---|--|-----------------------|
| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
| X | US 4 909 450 A (Henne et al.) 20 March 1990 see figures 1 to 7 | 1-2, 34 |

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/AU01/00062

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

| Patent Document Cited in Search Report | | | | Patent Family Member | | | |
|--|----------|----|----------|----------------------|----------|----|---------|
| GB | 2172586 | AU | 54395/86 | BR | 8601177 | CA | 1281907 |
| | | CN | 86101735 | DD | 250551 | DE | 3608899 |
| | | FI | 861077 | FR | 2582018 | IN | 165327 |
| | | IN | 168970 | JP | 61270342 | NZ | 215368 |
| | | SE | 8601005 | ZA | 8601957 | | |
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| | | AU | 52422/86 | CA | 1281185 | DD | 243939 |
| | | DE | 3603005 | NZ | 214910 | | |
| AU | 11627/83 | JP | 58152095 | US | 4508539 | | |
| US | 4909450 | DE | 3718781 | EP | 294609 | | |
| END OF ANNEX | | | | | | | |